Mariana Mascarenhas de Menezes Costenla

Licenciatura em Ciências da Nutrição

Heavy Metal Contamination in Seafood and Consumer Exposure in the Gulf Cooperation Council

Dissertação para obtenção do Grau de Mestre em Tecnologia e Segurança Alimentar

Orientador: Professora Doutora Benilde Mendes
Faculdade de Ciências e Tecnologia da Universidade Nova de Lisboa

Júri:

Presidente: Prof. Doutor(a)
Arguente(s): Prof. Doutor(a)
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ACKNOWLEDGEMENTS

Being able to finish this paper has given me an immense sense of accomplishment. Not only because it means that I have delivered what was asked for the completion of my Master´s degree, but also because of the challenge it implied. Defining the theme, the goals and conducting a research is challenging enough but doing so in a totally different country, in a different culture and a different language, Arab is not exactly intuitive and working full time in a company that was not so pleasant, away from all that is physically and emotionally familiar is even a greater challenge. The lack of local support and infrastructures for the development of my paper has made me change my strategy and follow a completely different approach. Instead of conducting a laboratory rehearsal with defined goals and methods, I had to look around to the society and find a problem for which I could create a hypothesis to then develop, based on bibliographic research. When all the dots finally aligned and I began this journey, it would be a matter of commitment, in which I succeeded. This was one of the most demanding periods of my life. It wouldn´t have been possible to achieve without the acceptance, the patience, the respect, the friendship, love and protection of Luisa and Andrew Rackham, who were my family, my friends, my confidents, my supporters, my everything to whom I cannot even describe the eternal gratitude and unconditional love that I feel. They received me with open arms into their house and lives and never for once made me feel unwelcomed. I had 4 sisters, 1 brother and 2 brothers in law. I now have 5 sisters, 1 brother and 3 brothers in law. What a family. Also would like to thank Veronica for making my life easier without ever complaining and Joana for listening every single day to my complaints giving me support, and for making it possible to unwind sometimes and enjoy good moments.

I would like to thank those who thought of me and never forgot I was still there. My dear friends, Ana MS, Rita RA, Maria S, Sofia M, Ana J, Zé Maria, Luís, André and Pedro, I know now with certainty that I was not mistaken and I feel blessed. But I must be fair to the technology available, thank you Google for having such a good video call system that allowed me to see my family every day making all the homesickness bearable.

Finally, I would like to thank my advisor, Professor Benilde Mendes, for encouraging me and never stop believing that I would be able to reach the end of this path.

RESUMO

A população nos países do Golfo tem vindo a aumentar e a pesca é uma das suas fontes de rendimento. A indústria instalada também tem aumentado e as descargas operadas no meio hídrico, particularmente pela indústria de dessalinização das águas e pelas refinarias leva à acumulação de metais pesados no ambiente e no pescado.

O objectivo deste trabalho é rever a presença de cádmio, arsénio, chumbo e mercúrio no pescado da Arabia Saudita, Oman, Kuwait, Emirados Árabes Unidos, Bahrain e Qatar e estimar o consumo humano destes metais nestes países.

Todas as amostras de marisco testado para o arsénio (As) mostraram concentrações acima dos valores máximos permitidos, e as provenientes do Qatar e dos EAU encontravam-se entre as mais contaminadas. Todas as amostras provenientes do Kuwait continham chumbo (Pb), cádmio (Cd) e mercúrio (Hg). As amostras do Bahrain continham Pb acima dos valores permitidos. Na costa de Oman e Qatar, o marisco continha Cd acima dos valores permitidos.

A ingestão semanal estimada (ISE) de As devida ao consumo de marisco está acima da ingestão tolerável semanal (ITS). Algumas amostras fornecem Cd, Pb e Hg acima do valor tolerável.

Nos EAU, existe peixe contaminado com Cd, na AS com Cd e Pb, nenhum ultrapassa a ITS. Os níveis mais altos de Hg foram encontrados no peixe do Kuwait e de As no Bahrain e Qatar. A ISE de As está acima da ITS para todo o peixe e a de Hg para o peixe dos EAU, Kuwait e Qatar.

A ingestão semanal de As e Cd pelo consumo de marisco é elevada. O peixe apresenta um perigo para a saúde pública devido ao As. Pode haver risco pela ingestão crónica de baixas doses. Os grupos de risco devem ser devidamente aconselhados.

Palavras Chave: Pescado, Metais Pesados, Golfo, Consumo Semanal Estimado

ABSTRACT

The population from countries surrounded by the Gulf and Arabian Sea depend on fisheries. Industry is growing and discharges by desalination plants and refineries lead to the presence of heavy metals which accumulate in the environment and seafood.

The aim is to review seafood contamination with cadmium, arsenic, lead and mercury in Saudi Arabia, Oman, Kuwait, United Arab Emirates, Bahrain and Qatar and estimate the consumer exposure to these metals.

All samples of molluscs and crustaceans tested for arsenic (As) showed concentrations above maximum permitted levels, Qatar and UAE being the most contaminated. All samples from Kuwait contained lead (Pb), cadmium (Cd) and mercury (Hg). Bahrain samples contained Pb above permitted levels. On the coast of Oman and Qatar shellfish showed concentrations of Cd above permitted levels. The EWI for As from shellfish is above the PTWI for all samples. For Cd, all except shellfish from Oman, Bahrain and KSA entail a EWI above the PTWI. None of the samples, except from Kuwait, provide Pb above the PTWI and for Hg, only the clam, mussel and crab contain higher amounts. Four locations in the UAE contained fish with Cd above permitted levels. Tuna fish from KSA had higher levels of Cd and Pb. Highest levels of Hg could be found in fish from Kuwait. Bahrain and Qatar showed the highest levels of As. Hg EWI exceeded for the UAE, Kuwait and Qatar.

Estimated weekly intakes of As and Cd from shellfish are extremely high and above the Provisional Tolerable Weekly Intake. Fish from these countries does not contain high levels that pose a threat to public health, except for As. The risk is probably in chronic low exposure. Risk groups should be properly advised.

Keywords: Seafood, Heavy Metal, Gulf, Consumer Exposure

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LIST OF ABBREVIATIONS

µg/g – microgram/gram
ANZFA – The Australia New Zealand Food Authority
As – Arsenic
BAPCO – Bahrain Petroleum Company
bw – Body weight
Cd - Cadmium
Co – Cobalt
Cr – Cromium
DHA – Docosahexaenoic acid
DMA (V) – Dimethylarsinic acid
DNA – Deoxyribonucleic acid
EC – European Commission
EDI – Estimated Daily Intake
EFSA – European Food Safety Authority
EPA – Eicosapentaenoic acid
EWI – Estimated Weekly Intake
FAO – Food and Agriculture Organization
GCC – Gulf Cooperation Council
GSO – Gulf Standard Organization
Hg – Mercury
IARC – International Agency for Research on Cancer
ISO – International Standard Organization
JEFCA – Joint FAO/WHO Expert Committee on Food Additives
KSA – Kingdom of Saudi Arabia
Mbd – Million barrels per day
Mean ± sd – mean ± standard deviation
MeHg – Methilmercury
MMA(V) – Monomethylarsonic acid
Ni – Niquel
OSG – Orange Spotted grouper
Pb – Lead
PTWI – Provisional Tolerable Weekly Intake
PUFA – Poliunsaturated Fatty Acid
ROPME – Regional Organization for the Protection of the Marine Environment
ROS – Reactive Oxygen Species
SBM – Single Buoy Morring
SE – Spangled emperor
UAE – United Arab Emirates
UV – Ultraviolet
WHO – World Health Organization

1. CONTEXT and AIM

Food is a substance that is capable of nourishing the human body with the adequate and necessary nutrients that support life, with plastic, energetic and regulative functions. We assume that food is safe to our health. But sometimes food contains not only the nutrients that provide good health but also other natural or introduced substances, often prejudicial. These are called contaminants. By definition, a contaminant is any substance not intentionally added to food, but which is present as a result of its production (including operations carried out in crop husbandry, animal husbandry and veterinary medicine), manufacture, processing, preparation, treatment, packaging, transport or holding or due to an environmental contamination. The term does not include insect fragments, rodent hairs and other extraneous matter. In case of heavy metals and other chemical components intentionally used by industries, contamination can occur through soil, water and air. Contamination of the environment is not only a pollution issue but also a rising public health matter, from the consumption of such contaminated food and water (World Health Organization, 2014). This monograph focuses on seafood and fish contamination by heavy metals captured and consumed in Gulf countries.

The Gulf is in constant expansion in the number of new industries and urban development. The tanker traffic is enormous and so is the probability of effluent discharge with unacceptable levels of heavy metals to the surrounding waters (Al-Jedah & Robinson, 2001).

I moved to Muscat, Oman during the period of the Master's degree after a work opportunity came across and I decided that it would be interesting to present my thesis on a different environment. My motivations on choosing this theme followed the horrific experience of visiting a fish market in Seeb, Muscat. With an outside temperature of around 40ºC fish was by the fishermen´s feet in open air, with absolutely no cooling system what so ever and covered in flies. The fish was bought from and handled by the same person that handled the money and then arranged in a different site, hot and humid, with no running water and hygienic conditions worthy of a Paris, 18th century, not that I've been there, but as described in the book "The Perfume" from Patrick Suskind:

"The streets stank of manure, the courtyards of urine, the stairwells stank of moldering wood and rat droppings, the kitchens of spoiled cabbage and mutton fat; the unaired parlors stank of stale dust, the bedrooms of greasy sheets, damp featherbeds, and the pungently sweet aroma of chamber pots. The stench of sulfur rose from the chimneys; the stench of caustic lies from the tanneries, and from the slaughterhouses came the stench of congealed blood. People stank of sweat and unwashed clothes; from their mouths came the stench of rotting teeth, from their bellies that of onions, and from their bodies, if they were no longer very young, came the stench of rancid cheese and sour milk and tumorous disease. The rivers stank, the marketplaces stank, the churches stank, it stank beneath the bridges and in the palaces. The peasant stank as did the priest, the apprentice as did his master's wife, the whole of the aristocracy stank, even the king himself stank, stank like a rank lion, and the queen like an old goat, summer and winter.

For in the eighteenth century there was nothing to hinder bacteria busy at decomposition, and so there was no human activity, either constructive or destructive, no manifestation of germinating or decaying life that was not accompanied by stench."

My initial idea was to evaluate the microbiology quality of fish bought and arranged in the market and compare it to the microbiology quality of fish arranged in the labs of Sultan Qaboos University. For that I met the Head of Department and Professor of Food Science and Nutrition, Dr Nejib Guizani and Dr Ismail Al Bulushi, Assistant Professor in the same department, College of Agricultural and Marine Sciences, who immediately agreed to help and co-work with me in this project but soon realised that authorization alone for carrying such analysis would take approximately 1 year.

Having to rely only on my own I looked around to the society. After a couple of experiences in the markets and realizing how poor the hygiene conditions fish caught was submitted too and determining the proximity of petroleum sites I decided to conduct a review on the heavy metal contamination of seafood followed by an estimation of the intake of these contaminants through fish from the surrounding areas. Being able to estimate this consumption could work as the basis to develop special recommendations on fish intake to specific groups in these countries.

The aim of this work is to review the presence of environmental contaminants in fish and seafood originated from the Gulf, Gulf of Oman and Arabian Sea and the consumer´s exposure to them within the Gulf Cooperation Council countries.

The specific aim of this work is to review heavy metal contamination, namely lead, cadmium, mercury and arsenic in fish and seafood from the Gulf, Gulf of Oman and Arabian Sea published between the year 2000 and February 2014 and estimate the weekly intake of such heavy metals by the consumer according to the apparent consumption of fish and seafood in these countries.

2. ENVIRONMENTAL CONTAMINANTS – HEAVY METALS

The presence of heavy metals in the environment has reached alarming dimensions. The discharges of residues by industry and above all, refineries lead to the contamination of soil and water by heavy metals (Bolana *et al.*, 2014). Though there is no precise definition for the term heavy metal, it can be defined by its density, which in this case is above $5 g/cm^3$. Heavy metals that pose more threat to human health are mercury (Hg), cadmium (Cd), lead (Pb) and arsenic (As) (Järup, 2003). Not only the industry but also coastal urban development allows large quantities of metals to be released in the marine environment through discharge of waste waters. Unlike other organic compounds, metals do not suffer chemical or biological degradation, so their concentration in either soil or water can remain at very high levels even after long periods of time after their introduction in the habitats (Adriano *et al*., 2004).

Authorities define not only the maximum permitted levels of heavy metals in marine biota but define as well the provisional tolerable weekly intakes (PTWI) for humans (Table 2.1).

Table 2.1: Provisional Tolerable Weekly Intake (PTWI, µg/Kg body weight) for heavy metals in humans and maximum accepted levels (mg/kg wet weight) in fish and marine biota

Reference	Cd		Pb		Hg		MetilHg*		As	
	PTWI	**Fish**	**PTWI**	**Fish**	**PTWI**	**Fish**	**PTWI**	**Fish**	**PTWI**	**Fish**
(JEFCA, 2003)							1.6	0.5 – 1.0		
(Comissão Europeia, 2006)		0.05-1.0		0.3 - 1.5		0.5 - 1.0				
(EFSA, 2009) (EFSA, 2010) (EFSA, 2012)	2.5		25		4					
(JECFA, 2010)					4				0.3 - 8.0	
(ANZFA, 2013)		2.0[a]		0.5 – 2.0		0.5 – 1.0				1.0 - 2.0

* - will be used as reference for the calculation of the EWI; a – molluscs only

2.1 Source and toxicity

2.1.1 Mercury

Mercury (Hg) is distributed in the environment and is non-essential and toxic to the human body. Methylmercury compounds are possibly carcinogenic to humans, Group 2B of IARC classification (WHO, 1993). Mercury pollution of the environment has natural, anthropogenic, and historic sources, has doubled the last century and is mainly a man-made problem. Anthropogenic mercury is released from numerous sources being fossil burning one of them (Bose-O'Reilly *et al.*, 2010). It is considered to be one of the major environmental pollutants, is widely used in industry, agriculture and medicine, and circulates in the ecosystems, but it is never destroyed and cannot be eliminated by cooking (Bose-O'Reilly *et al.*, 2010).

Chemically, mercury exists in various forms as elemental mercury, inorganic mercury and organic mercury compounds. Methylmercury (MeHg) and ethylmercury are common organic forms of mercury combined with carbon. Methylmercury is also formed from methylation of inorganic mercury by microorganisms in the environment and constitutes the major source of exposure from seafood. Humans are usually exposed to mercury in a chronic and low dose (Park & Zheng, 2012). Mercury in contaminated water has the potential to enter the food chain and once in the food chain, it bioaccumulates causing adverse effects to human health (Rice *et al.*, 2014). Fish, from fresh or salt water, appear to be the primary source of MeHg poisoning in humans (Bernhoft, 2012). Through mechanisms which are not yet known, various species of fish tend to have higher rates of MeHg bioaccumulation (Rice *et al.*, 2014). MeHg levels tend to be higher in large predators (e.g., shark, swordfish, king mackerel, tilefish); intermediate in medium-sized predators (e.g., trout, snapper); and lower in short-lived (e.g., salmon) or smaller (e.g., shrimp, clams) species (Mozaffarian & Rimm, 2006).

Once ingested the gastrointestinal tract absorbs approximately 95% of MeHg. Because urinary excretion of MeHg is negligible, MeHg is primarily eliminated from the body in an inorganic form through the action of the biliary system at the rate of 1% of the body burden per day. Approximately 90% is excreted in stool and 20% of methyl mercury is excreted in breast milk from lactating women (Bernhoft, 2012). The biological half-life of MeHg is 39 to 70 days depending on body burden (Guzzi G, 2008). Once MeHg is ingested, it is absorbed through the epithelial cells and can cause various digestive disturbances like inhibiting the production of the digestive enzymes (Vidjani *et al.*, 2003).

Human toxicity varies with the form of mercury, the dose and the rate of exposure (Bernhoft, 2012). Mercury can be quickly removed from the blood and redistributed into different tissues. It is thought that shortly after entering the body it becomes bound in the brain, spinal cord, ganglia, autonomic ganglia, and peripheral motor neurons. Although the nervous system is the primary repository for mercury exposure, the systemic distribution has the potential to cause symptoms in different organ systems (Gundacker, 2010) causing cellular, cardiovascular, haematological and pulmonary effects, effects on digestive and renal system, on the endocrine, reproductive system and also affect the foetus (Rice *et al.*, 2014).

Mercury exposure is associated with oxidative stress, increased radical oxygen species and DNA damage (Flora *et al.*, 2008). Some studies demonstrated that MeHg can inhibit the cardio protective activity of paraoxonase 1, may be a cause for anaemia and be involved in leukaemia (Pyszel *et al.*, 2005). It is related to increased blood pressure, risk of myocardial infarction, atherosclerosis and risk of developing cardiovascular disease (Azevedo *et al.*, 2012). In the gastrointestinal system the effects of mercury may be such as abdominal pain, indigestion, inflammatory bowel disease, ulcers and bloody diarrhoea. It has also been associated with the destruction of intestinal flora and reduced resistance to pathogenic infection. It can also cause kidney damage to a large extension (Miller *et al.*, 2013).

Mercury can produce an immune response in the central nervous system and induce alterations in immune cell production and function, creating an individual chronically susceptible to infection, chronic disease or even auto immune conditions (Gardner *et al.,* 2010). Mercury accumulates in the nervous tissues throughout the body (Ceccatelli *et al*., 2010). It can damage the blood brain barrier and it facilitates penetration of the brain by other toxic metals and substances, causing effects in the central nervous system and also immune, sensory, neurological, motor, and behavioural dysfunctions.

Low exposure levels of mercury may affect the endocrine system by the disruption of the pituitary, thyroid, adrenal glands and pancreas (Minoia *et al*., 2009). The effects on the reproductive system affect not only conception but also the foetus development. Methil-Hg can cross the placenta and cause neurological damages to the foetus and babies can be born with a variety of birth defects and cognitive delays (Bernhoft, 2012; Rice *et al.*, 2014). It is also excreted into breast milk jeopardizing the baby´s health. Nevertheless, the advantages of breastfeeding outweigh the possible risks of exposure. Children are considered especially vulnerable to these environmental threats; exposures are often higher due to body weight. There are specific periods in their development when the exposure to a chemical, physical, or biological agent may result in adverse health. Women who may become pregnant, already pregnant women, nursing mothers, and young children should avoid some types of fish and eat fish and shellfish that are lower in mercury like those in a lower position in the food chain (Bose-O'Reilly *et al.*, 2010).

Apart from the mercury contamination, fish and seafood have nutritional benefits.

2.1.2 Cadmium

Cadmium (Cd) is one of the most toxic elements to which humans are exposed. It is classified as a human carcinogen for which the mechanism of action is unknown. The IARC classification is Group 1 which means "Sufficient evidence in humans or sufficient evidence in animals and strong mechanistic data in humans" (WHO, 2012).

Environmental exposure mainly occurs by contact with tobacco smoke, water and food. By far the highest concentrations are found in fish and shellfish, the highest contributor of Cd for non-smokers (Llobet *et al.*, 2003). Cd is not well absorbed; the inhalation, oral and dermal routes are associated with absorptions of 25%, 1–10%, and <1%, respectively (Borchers *et al*., 2010). It is widely distributed throughout the body, but the highest levels are found in liver and kidney. It is not known to undergo any metabolic conversion and is eliminated very slowly, with 0.007% and 0.009% of total body burden being excreted daily via urine and faeces, respectively (Borchers *et al*., 2010). This metal gradually accumulates in the body and levels increase with age given its long half-life, which is more than 20 years (Sabath & Robles-Osorio, 2012).

Recent studies suggest that Cd exposure may result in an altered DNA methylation pattern (Cheng *et al*., 2012). Evidence suggests that exposure to cadmium does not induce direct DNA damage; however it induces an increase in reactive oxygen species (ROS) formation, which in turn induces DNA damage and can also interfere with cell signalling. More important seems to be cadmium interaction with DNA repair mechanisms. It is of particular importance that these effects are observed at exposure to low, micromolar or even sub-micromolar cadmium concentration that may be relevant to human exposure (Filipic, 2012).

Cadmium is able to pass to the foetus via the placenta affecting its central nervous system. It can impair myelination in the neonate, create deficits in learning and altered behaviour. Increasing evidence has demonstrated that Cd is a possible etiological factor of neurodegenerative diseases, such as Alzheimer's disease and Parkinson's disease (Wang & Du, 2013).

Various epidemiological studies have demonstrated that environmental exposure to Cd increases the risk of developing kidney injury (Sabath & Robles-Osorio, 2012). High levels of Cd in urine were associated with higher overall mortality and increased risk of cancer (Menke *et al*., 2009). In addition to its nephrotoxic effect, Cd is also associated with increased risk of developing diabetes (Schwartz *et al*., 2003). Exposure to Cd also increases the risk of high blood pressure and it is considered a risk factor for cardiovascular mortality and morbidity (Gallagher & Meliker, 2010). In addition to clinical cardiovascular outcomes, chronic exposure to cadmium has also been associated with cardiovascular disease risk factors and with subclinical events, such as hypertension (Tellez-Plaza *et al*., 2013).

Prevention is the most important factor in the management of exposure to this metal, since there is no effective means of treating Cd toxicity (Sabath & Robles-Osorio, 2012).

2.1.3 Lead

Lead (Pb) is a soft and malleable metal, which is considered a heavy metal. It is a public health problem due to its adverse effects, mainly affecting the central nervous system in the most vulnerable populations, such as pregnant and lactating women and children (Koyashiki *et al*., 2010). It´s IARC classification is Group 2A, "Limited evidence in humans and sufficient evidence in animals" (WHO, 2006).

Once it enters the blood stream, 99% of Pb binds with the erythrocytes (Shafiq-ur-Rehman, 2013). After it reaches the bloodstream it has a half-life of roughly 30 days and is excreted in urine and bile. The remaining Pb binds to red blood cells, is distributed throughout the soft tissues of the body and accumulates in the bone. Once in the bone, its half-life ranges from 20 to 30 years. Events like pregnancy, menopause or lactation increase the turnover of bone tissue and releases lead back into the bloodstream (Mason *et al*., 2014).

During the pregnancy, the level of lead in maternal bloodstream increases, it crosses the placenta which poses a threat to the health of the foetus. Deficiencies in calcium, iron, and zinc have been identified as risk factors for toxic effects being calcium deficiency one of the main causes for the severity of the effects (Mason *et al*., 2014). Children are at higher risk because they have greater gastrointestinal absorption and less effective renal excretion and their encephalic barrier is immature (Koyashiki *et al*., 2010). Not only during the pregnancy but also during lactation, Pb can pose a threat to infants. Breast milk from mothers with current exposure to lead or mothers exposed by the redistribution of bone lead has been identified as a source of exposure to the infant (Koyashiki *et al*., 2010).

The presence of Pb in the human body causes damage to the nervous system through several mechanisms. Neuropsychological research over the years has revealed that Pb exposure can result in declines in intelligence, memory, processing speed, comprehension and reading, visuospatial, motor and executive skills. Among the cognitive deficits induced by Pb toxicity, visuospatial deficits appear to be major. Anxiety, depression and phobia can also occur, while outcome, intervention, and rehabilitation results are largely dependent on the level of toxic exposure. There is also a growing evidence of antisocial behaviour linked to early Pb exposure (Mason *et al*., 2014).

Research also indicates that cumulative environmental Pb exposure is neurotoxic to adults acting as a risk factor for accelerated declines in cognition; it is proposed that there might be an association with late onset of Alzheimer’s disease, but this relationship as not yet been established (Bakulski *et al*., 2012). It is also proposed that it might work as a risk factor for the expression of schizophrenia later in life, alone or in combination with other environmental factors during exposure in foetal or neonatal life (Guilarte *et al*., 2012).

Oxidative stress have already been defined has a mechanism for toxicity of Pb. It has been reported that it induces degradation of the membrane phospholipids in the brain and increases lipid peroxidation, which in turn can be influenced by the presence of pro or anti-oxidant systems with either prejudicial or beneficial effects (Shafiq-ur-Rehman, 2013).

2.1.4 Arsenic

Arsenic (As) is a metalloid that is naturally present in low concentrations in the environment (Rodríguez-Barranco *et al.*, 2013). It can be categorised as organic or inorganic, depending on the presence or absence of a carbon bond and may be found in one of three oxidation states, −3, +3 and +5, the trivalent form being the most toxic (Orloff *et al.*, 2009). The toxicity of the pentavalent form results from its conversion to the trivalent form (Jomova *et al.*, 2011). The inorganic forms of arsenic are generally more toxic than the organic forms and are responsible for most cases of arsenic poisoning in humans. Arsenic is a well-established human carcinogen (Bhattacharjee *et al*., 2013). It´s IARC classification is Group 1, “Sufficient evidence in humans or sufficient evidence in animals and strong mechanistic data in humans” (WHO, 2012). The health consequences of arsenic exposure include respiratory, gastrointestinal, haematological, hepatic, renal, skin, neurological and immunological effects, as well as damaging effects on the central nervous system and cognitive development in children (Argos *et al*., 2010; Rosado *et al*., 2007).

The main routes of exposure to inorganic arsenic are ingestion of drinking water and inhalation of polluted air and dust, the former being the most important route in the case of millions of children in countries with high levels of arsenic in water (Smedley & Kinniburgh, 2002). The organic compounds normally present in food, especially fish and seafood, are considered nontoxic or of low toxicity (Stea *et al*., 2014). Organic arsenic, in turn, is mainly found in fish and seafood in the form of arsenobetaine and arsenocholine (Orloff *et al*., 2009).

After ingestion, about 60–90% of organic and inorganic forms alike are absorbed into the bloodstream from the gastrointestinal tract (Hall, 2002). Arsenic needs to go under methylation as part of its biotransformation which was previously considered detoxification, but is now known that the metabolite it produces is more toxic than the form exposed to previously. Monomethylarsonic acid MMA(V) and dimethylarsinic acid DMA(V) are the end products of arsenic metabolism. These are excreted through the urine. This balance between intake and excretion will determine the load of arsenic in the system (Bhattacharjee *et al*., 2013).

One of principal mechanisms of arsenic toxicity is the induction of a strong oxidative stress with production of free radicals in cells that induce DNA damage, lipid peroxidation and decreased glutathione levels. The oxidative stress induced by chronic exposure to inorganic As is related to cytotoxic and genotoxic effects in the cells, acting as cause in the pathogenesis of diabetes, cardiovascular and nervous systems disorders. The inhibition of DNA repair processes is considered the main mechanism of genotoxicity (Faita *et al*., 2013). Cancer in skin, liver and kidney may originate from the molecular damage to proteins, lipids and DNA (Jomova *et al.*, 2011).

The extent of arsenic poisoning depends on various factors such as dose, individual susceptibility and the age of the affected individuals (Jomova *et al.*, 2011).

Arsenic can be transferred from the mother to the foetus through the umbilical cord. That could create some adverse birth outcomes, increase infection and increasing the risk for neurobehavioural impairment, cardiovascular disease and high blood pressure during childhood (Farzan *et al*., 2013). The developing human brain is especially vulnerable to toxic chemical exposures during pregnancy, infancy and childhood with more permanent damaging effects with levels of exposure that would have little effect in an adult (Grandjean & Landrigan, 2014).

While the influence of in utero or early life As exposure on neurotoxicity may impact the risk of chronic disease, the long-term consequences of these early alterations have yet to be elucidated (Farzan *et al*., 2013). These studies refer to children and mothers exposed to arsenic from water supplies. Nevertheless, even from seafood consumption children and infants may have higher exposure to metals because they consume more food in relation to their body weight and absorb metals more readily than adults (Horton *et al*., 2013).

Some studies have linked high and low levels of As exposure to type 2 diabetes mellitus but mostly from drinking water, tap water, ground and well water. Further studies are needed in order to better understand the role of As in the development and progression of this type of diabetes (Andra *et al*., 2013).

High levels of As exposure in drinking water have also been related to increased risk of cardiovascular disease (Stea *et al*., 2014). The association is mostly with peripheral arterial disease and coronary heart disease. This association is partly independent of traditional risk factors but it is still necessary to conduct more studies in order to assess the dose, the risk and the individual susceptibility to the occurrence of a cardiovascular disease. It is also suggested that there is an association between arsenic exposure and the prevalence of hypertension (Abhyankar *et al*., 2012).

There is ample evidence of the renal damage associated with these heavy metals. In fact, the combination of different metals has been shown to have a cumulative nephrotoxic effect.

Most of the evidence lacks variables necessary to assess dose-response causality and evidence in humans is not so well established given the small number of studies with both high and low level exposures to both inorganic and organic arsenic.

3. THE COOPERATION COUNCIL FOR THE ARAB STATES OF THE GULF

On the 25th of May 1981, the 21st Rajab[1] 1401 of the Islamic calendar, the leaders of the United Arab Emirates (UAE), the State of Bahrain, Kingdom of Saudi Arabia (KSA), Sultanate of Oman, State of Qatar and State of Kuwait, signed a deal of cooperation between the six countries in order to unify and strengthen business and areas of cooperation between its citizens and to stand for the goals and interests of the Arab nation respecting the common principles of Islam (Figure 3.1). This decision was based upon the strong religious and cultural beliefs as well as the close geographic position between these countries, which assures the communication and interaction amongst them.

The development of "The Cooperation Council for the Arab States of the Gulf" (GCC) was therefore able to unify and upgrade the response in the areas of safety, health and economic growth including education, culture, information and tourism, legal, commercial and social affairs. Another area of great importance is the area of science and technology related to marine and agricultural resources (The Cooperation Council for the Arab States of the Gulf, 2012a). The Council is composed by the representatives of each state and the Head of it changes periodically respecting the alphabetical order of the name of the country (The Cooperation Council for the Arab States of the Gulf, 2012b).

Figure 3.1- GCC map– Kingdom of Saudi Arabia, Sultanate of Oman, United Arab Emirates, State of Qatar, State of Bahrain and State of Kuwait (adapted from www.maps.google.pt, search: Middle East)

[1] Rajab is the 7th month of the lunar calendar. The first year coincided with the year 622 CE (Common Era = After Christ). The lunar calendar starts immediately with the sunset and the sighting of the new moon, process that depends on several factors as weather conditions.

According to Article 1, chapter 1 of the Economic Agreement of December 31st 2001, all foods produced by one member of the GCC are treated as national produce by other members of the GCC (The Cooperation Council for the Arab States of the Gulf, 2004). These countries have recently become aware of the urgent need to develop efficient food control infrastructures, capable of providing the adequate protection for the consumer. Effective national food control systems are essential to ensure the safety of food and to protect consumers given the lack of control systems which then lead to a higher incidence of foodborne diseases. They are also critical in enabling countries to assure food safety and quality entering international trade and to ensure that imported foods conform to national requirements (Food Regulation Middle East, 2012a).

The Gulf Standards Organization (GSO) is comprised of senior standards officials from the six GCC member countries adding to a representative from Yemen. GSO is responsible for developing food and non-food standards in the GCC. The GSO food standards committee has been working over the past few years to harmonize existing standards within the guidelines of the *Codex Alimentarius*, ISO and other international organizations (Food Regulation Middle East, 2012b). The role of GSO's food safety policy is the protection of the health and safety of all people in GCC through the maintenance of a safe food supply via prevention of food borne diseases, prevention of chemical hazards and food contamination at any stage of the food chain. It applies to all countries.

3.1 GCC countries – Geography, Industry and Fisheries Information

As previously mentioned, the GCC is formed by 6 countries such as Saudi Arabia, Kuwait, Bahrain, Oman, Qatar and the UAE. Geographic and demographic characteristics of each country are presented in Table 3.1 (GCC-SG, 2012a) (FAO, 2014a).

Table 3.1 - Demographic and geographic characteristics by country (GCC-SG, 2012a; FAO, 2014a)

Country	Capital	Population (million)	Area (1000km2)
Saudi Arabia	Riyadh	28.705	2000
Bahrain	Manama	1.359	0.767
Kuwait	Kuwait City	2.892	17.8
Oman	Muscat	2.904	309.5
UAE	Abu Dhabi	8.106	71
Qatar	Doha	1.939	11.6

The geographic position of these countries is of extreme importance for the contextualization of this paper. These countries have coasts facing the Persian Gulf which continues to the Gulf of Oman and the Arabian Sea. Connecting both Gulfs is the Strait of

Hormuz, a very narrow passageway with only 36 meters of depth, through which all shipments heading or leaving the Gulf cross (Kosanovic *et al.*, 2007). Annually around 25,000 tanker movements sail in and out of the Strait of Hormuz and transport about 60% of all the oil carried by ships. The Gulf is located in a subtropical, hyper-arid region and has a maximum depth little no more than 60 meters. Its photic zone extends to only 6–15 m (Sheppard *et al.*, 2010). The Arabian shore is mostly sedimentary and comprises shallow, semi-enclosed sea with very high evaporation rates and poor flushing characteristics which contributes to a slower dispersion of contaminants in a constant high temperature, high salinity and UV exposure (Mora *et al*., 2004). The general lack of precipitation, high temperatures in summer and dry winds during winter create 1–2 m equivalent of evaporation per year (Sheppard *et al.*, 2010) and these characteristics create a stressed and fragile marine environment.

These waters are heavily utilised, since they are the pathways for tanker traffic to, from and between the GCC countries, with the consequential discharges related to shipping activities (Mora *et al*., 2004). Adding these ship movements to an increase in the surrounding industrial and urban developments, the number of effluent discharges reaching the sea with unacceptable levels of heavy metals is of great concern (Al-Jedah & Robinson, 2001). Desalination plants also contribute for this stressed environment. This industry returns to the Gulf over 7 km^3/year (numbers from 2005) of brine which is commonly hot and often contains pollutants that come from the pipes. Extreme aridity and unfavourable geology result in not having suitable alternative sources of potable water for domestic and industrial use therefore needing to use sea water for human consumption (Sheppard *et al.*, 2010).

The Regional Organization for Protection of the Marine Environment (ROPME) Sea Area which comprises the Gulf and Gulf of Oman was created to coordinate the Member States efforts towards protection of the water quality in this area and protect the environment systems as well as marine living and to abate the pollution caused by the development activities of the Member States (ROPME, 2013). It has undergone several surveys and changes but despite its efforts there is still limited exchange of information among government agencies, projects or neighbouring countries (Sheppard *et al.*, 2010).

The political context is also of great importance as it sometimes leads to episodes of conflict such as the Gulf War in 1991 following Iraq´s invasion of Kuwait, where there was a conflagration of over 700 oil wells that burned for months and an intentional discharge of 6-10 million barrels of crude oil into the marine environment, the largest oil spill in history (Al-Hassan *et al*., 2003). This reached over 770 km of coastline from southern Kuwait to Abu Ali Island (Saudi Arabia), with oil and tar erasing most of the local plant and animal communities (Hussain & Gondal, 2008).

If not by intention, but by natural causes, this area is a hot spot for pollution. It is also of great industrial development following the exploration of its natural resources. Last data indicates that the GCC alone contained 4% of the world oil reserves and is responsible for the production of 15 million barrels of oil per day (mbd) against a total of 19 mbd from all the other countries in the world. Regarding natural gas, the GCC contains 22% of the world natural gas reserves (GCC-SG, 2012b).

Population in these countries has increased by 50% between the years 2000 and 2012 (FAO, 2014a). As the population increases, so does the demand for food. Next, there is a description on the consumption of fish and seafood, including only fish, crustaceans and molluscs, per country of the GCC according to statistical data available. Latest FAO statistics estimate the apparent consumption per capita according to an average of consumption data provided between the years 2006 and 2010.

3.1.1 United Arab Emirates

Fisheries statistics, dated last from 2011, stated a total of 75174 tons of fish and seafood caught of which 25622 were fish and 1411 tons were molluscs and crustaceans and a total number of 24765 fishermen and 6370 fishing boats (UAE-NBS, 2013). FAO reports a total of fish production, including capture and aquaculture, of 75.2 thousand tons in 2011 (FAO, 2014b).

The UAE not only imports fish, crustaceans, molluscs and aquatic invertebrates but also exports. Last data, also from 2011, considers 15131 tons of exported seafood (UAE-NBS, 2012). In 2010, the average apparent consumption of fish and fish products in the UAE was of 28.6 kg/per capita (FAO, 2013). This represents approximately 548.1g/ per capita/ week.

3.1.2 Kingdom of Saudi Arabia

According to FAO´s latest Fishery and Aquaculture Statistics, in 2010, the population of Saudi Arabia had an average apparent consumption of fish and fish products of 8.2 kg/per capita (FAO, 2013). This represents approximately 156.8g/ per capita/ week. Data from FAO´s report on Food and Agriculture in the Near East (2014) reports a total fish production, including capture and aquaculture, of 90.8 thousand tons in 2011 (FAO, 2014b).

3.1.3 State of Qatar

FAO reports a fish supply of 24.5 kg/per capita (FAO, 2013) with a total production, including capture and aquaculture, of 13 thousand tons in 2011 (FAO, 2014b). The apparent consumption is approximately 469.7g/ per capita/ week.

3.1.4 Sultanate of Oman

FAO reports a fish supply of 28.7 kg/per capita (FAO, 2013) with a total production, including capture and aquaculture, of 159.2 thousand tons in 2011 (FAO, 2014b). The apparent consumption is approximately 550.2g/ per capita/ week.

3.1.5 State of Kuwait

FAO reports a fish supply of 17.9 kg/per capita (FAO, 2013) with a total production, including capture and aquaculture, of 4.4 thousand tons in 2011 (FAO, 2014b). The apparent consumption is approximately 343g/ per capita/ week.

3.1.6 State of Bahrain

FAO reports a fish supply of 13.5 kg/per capita (FAO, 2013) with a total production, including capture and aquaculture, of 10 thousand tons in 2011 (FAO, 2014b). The apparent consumption of fish is approximately 259g/ per capita/ week.

4. DISTRIBUTION OF HEAVY METALS IN ALGAE, SEDIMENTS, FISH AND SEAFOOD IN FROM THE GULF TO THE OMANI COAST OF THE ARABIAN SEA

Over the last two decades, much concern has been expressed regarding the state of the health of the water bodies in the Gulf region, given the rapid rate of growth and industrialization taking place there, the disproportionate amount of tanker traffic and the war-related events in the northern sector which have led to major sources of coastal marine pollution (Fowler *et al.*, 2007). The presence of heavy metals in several types of organisms, from plankton to fish, and also sediments, have been studied for the last years as there is a tendency to an increase of its concentrations, which suggests contamination from anthropogenic sources. Most of the studies identify zones as hotspots for pollution by heavy metals and they are all connected to areas of intense industrial activity (Naser, 2013). These metals accumulate in the bodies of marine biota at concentrations much higher than those found in the ambient water and are biomagnified in the food chain at higher trophic levels, posing a risk to human consumers (Saka *et al*., 2006). Data concerning either the presence of heavy metals in fish and seafood or the influence of the amount of seafood consumption on the levels of heavy metals in the human body is insufficient.

The locations where algae, sediments, molluscs or fish samples were collected along the coasts of the Gulf, the Gulf of Oman and the Arabian Sea are represented by red dots in Figure 4.1.

Figure 4.1 - Locations where algae, sediments, molluscs and fish were collected along the countries of the GCC (adapted from www.maps.google.pt, search: Middle East)

4.1 Algae and Sediments

Plankton and algae are included in the food chain. Algae are part of some fish´s natural food preferences, which are consumed by humans, but also by some bigger fish which will also be consumed by humans. For their characteristics, algae have been utilized as bio indicators for heavy metal pollution along the Gulf coast (Naser, 2013).

Heavy metals have been detected in algae along the coast of Saudi Arabia. In 2006, Al-Homadain tested 4 species of algae and detected concentrations of Pb between 8.84-18.60 µg/g (Al-Homadain, 2006). In 2007, the same author tested 3 different species of algae (*Chaetomorpha aerea, Enteromorpha clathrata* e *Ulva lactuca*) and found concentrations between 13.90 and 30.50 µg/g of Pb (Al-Homaidan, 2007). The author found a rise in the concentration of Pb in the algae and attributed this rise to the rapid industrialization and urbanization of the area, including the effects of oil pollution. In Dhofar, the southern part of Oman, Fowler and his collaborators decided to assess a temporal variation between algae contamination of heavy metals (Fowler *et al.*, 2007). The authors tested brown, green and red algae for Hg, Pb and Cd in 2002, compared it to values found for the same species in a survey carried out in 1983 and found both levels of Hg and Pb to have decreased and an increase for Cd levels, which given the small anthropogenic influence of the area, could be attributed to the geographic characteristics of the area. Even so, the authors state the difficulty in establishing a consistent temporal trend based on two surveys spaced 19 years apart (Fowler *et al.*, 2007).

Sediments are also a great indicator of environmental pollution. Heavy metals that are conveyed by sedimentary transport are largely retained within near shore regions. Heavy metals accumulated in bottom sediments may be released by various processes and enter the biologic chain, which in turn moves up to the food chain reaching humans (Al-Darwish *et al*., 2005).

The most recent studies carried out in the coastal regions of the GCC countries, with the highest values for metals, are highlighted (Table 4.1).

Al Darwish and collaborators (2005) studied the presence of heavy metals along the coastal region of Dubai in 2002. Amongst all areas, the areas Al-Hamriya and Dry Docks showed the highest levels of Pb, 35 and 20 ppm respectively and represent the two areas that are most contaminated, regarding other metals tracked by the authors. This area may be that contaminated since Al Hamriya Port is part of navigation routs. Also the staining of wooden ships may as well contribute to the higher levels of these metals and as well Dry Docks exhibit high levels of Pb which may be in part from the repairing and painting of different kinds of ships (Al-Darwish *et al*., 2005).

Another paper, by Mora and collaborators (2004) evaluated the presence of metals in the coastal area of Oman, Qatar, UAE and Bahrain. This work collected samples from several regions during 2000 and 2001 and encountered two major contaminated areas, the area surrounding BAPCO (Bahrain Petroleum Company) in Bahrain and Akkah Beach on the eastern coast of UAE. Regarding the sediments collected along the BAPCO refinery, the authors found

99 µg/g of Pb, 0.220 µg/g of Hg and 0.182 µg/g of Cd which most likely come from the industrial complex. Akkah Beach on the other hand is not a populated area, nor has any relevant industries close by, but still has high levels of Co, Ni and Cr, which are probably due to the local mineralogy and geology (Mora *et al*., 2004). In Oman, high levels of Cd were found in Raysut Port Area and Mughsayl, 0.20 and 0.21 µg/g respectively even higher than in BAPCO area in Bahrain. Mercury levels were relatively low in all regions apart from BAPCO with 0.22 µg/g which exceeds the sediments quality guideline value of 0.15 µg/g (Long *et al.,* 1995). Regarding As, the highest value was found on Akkah Beach, 9.6 µg/g which also exceeded the set limits of 8.2 µg/g (Long *et al.,* 1995).

Sadiq (2002) evaluated the presence of metals in an outfall area of a desalination plant in Saudi Arabia. Even though the highest values were found on the immediate vicinity of the outfall, some remained high at a much faraway location. Cd and Hg tend to dilute with the distance, but Pb on the other hand does not follow this trend (Sadiq, 2002).

Table 4.1 - Heavy metal presence (µg/g) in sediments

Country	**Area**	**Metals**				**Reference**
		Pb	**Cd**	**Hg**	**As**	
Saudi Arabia	Ras Tanajib	46.33	31.59	0.15		Sadiq, 2002
UAE	Dry Docks	20*				Al-Darwish *et al*., 2005
	Al-Hamrya St 3	35*				Al-Darwish *et al*., 2005
	Akkah Beach	1.33	0.09		9.6	Mora *et al.*, 2004
	Al Ruweis	5.88	0.11		2.2	Mora *et al.*, 2004
Oman	Mina Al Fahal	1.59	0.16		5.01	Mora *et al.*, 2004
	Raysut Port	0.729	0.20		1.09	Mora *et al.*, 2004
	Mughsayl	0.253	0.21		0.74	Mora *et al.*, 2004
Bahrain	BAPCO	99	0.18	0.22	4.96	Mora *et al.*, 2004
Qatar	Dukhan	3.88	0.07		6.3	Mora *et al.*, 2004
	Doha	3.16	0.08	0.01	5.0	Mora *et al.*, 2004

* - ppm

4.2 Molluscs, bivalves and crustaceans

Seafood is one of the biggest economic resources for the population in the GCC. Many studies have evaluated the concentration of some metals in oysters, clams, shells and crabs, food we would normally see in a fish market. Molluscs and bivalves have also been used as bio monitors to detect the presence of heavy metals in marine environment in the Gulf. They are sedentary animals that are easily exposed to environmental pollution as they usually live in shallow waters (Al-Busaidi *et al.*, 2013).

In the Eastern coast of Saudi Arabia, in the Arabian Gulf, Al-Saleh and Al-Doush (2002) evaluated the level of Hg in the shrimp *Penaeus semisulcatus* and found concentrations, mean ± sd, between 0.012±0.001 and 0.039±0.012 µg/g wet weight (Al-Saleh & Al-Doush, 2002). In the same coast, but in a different location and more recently, clams of the species *Meretrix meretrix* were evaluated for the concentration of Cd, maximum mean ± sd of 0.90±0.030 µg/g wet weight, and Pb, 2.49 ± 0.27 µg/g wet weight (Alyahya *et al*., 2011). Even though the Cd concentration did not exceed the maximum permitted levels by the European Community (EC), values found were extremely high. Regarding the Pb levels, they clearly exceeded the EC limits of 1.5 µg/ g wet weight for molluscs (Comissão Europeia, 2006).

In 2004 Mora and his collaborators published the results of the presence of heavy metals in pearl and rock oysters as well as pen shells and clams along the coast of the Gulf and Gulf of Oman. Concentrations were determined in dry weight. The highest concentration of Pb, 3.92 µg/g, was found in pearl oysters off the BAPCO refinery in Bahrain, followed by Abu Dhabi, with 2.29 µg/g. The highest value for Hg, 0.31µg/g, was found in clams from Ras Al Nouf in Qatar, followed by 0.20 µg/g in pen shells in Jebel Ali, Dubai. As for the Cd concentration, the highest values were found in Oman, 21.9 µg/g and 19.9 µg/g, in rock oysters in Ras Al Yey and Al Sawadi, respectively. Arsenic came to be one of the worrisome metals with the highest concentration in Qatari clams, 156 µg/g, as well as in pen shells from Abu Dhabi, 153 µg/g (Mora *et al.*, 2004). Apparently, there is no other data concerning As levels in molluscs and bivalves from the ROPME Sea Area, which makes it a matter of concern and probably a need to check the factors that contribute to these values (Mora *et al.*, 2004).

In 2005, Musaiger and Al-Rumaidh tested crabs from the coast of Bahrain. The authors tested raw and traditionally cooked crabs and found that levels of contamination decreased with cooking. In cooked crabs they found concentration levels, in mean ± sd, of 0.035 ± 0.0175 mg/100g wet weight for Pb and Hg and 0.0575 ± 0.0315 mg/100g wet weight for Cd, all within EC levels (Musaiger & Al-Rumaidh, 2005). Also in Bahrain Al Sayed e Dairi (2006) evaluated the concentration of metals in the snail of the species *Turbo coronatus* and found maximum levels of Pb of 3.03 µg/g wet weight, higher than the permissive levels set by the EC (Al-Sayed & Dairi, 2006).

In Kuwait Tarique and collaborators (2012) evaluated the organs of the *Amiantis umbonella* to monitor metal contamination in coastal sediments. These clams were caught in Kuwait bay near Al Doha in a contaminated site near a desalination plant, and from a reference site 5 km away. Generally, clams from the contaminated site had higher concentrations of metals than clams from reference site, which is not of surprise. The total metal concentration, in mean ± sd, for the whole clam from the contaminated site is of 11 ± 1,2 µg/g of Hg, 107.1±1.8 µg/g Cd and 163.1±4.2 µg/g wet weight of Pb. As for the reference site the concentrations were, for Hg, Cd and Pb, 1.5±0.9 µg/g, 34.7±1.7 µg/g and 45.3±2.5 µg/g wet weight respectively. The authors found that not only the distribution of metals among the organs reflect the availability of the metals in the environment but also that this specific clam accumulates Cd and Pb mostly in their kidneys and Hg was evenly distributed among the organs. Despite the fact that metals are distributed and accumulate more in some organs than others does not reckon any particular interest for us as consumers, since we eat the whole body of the clam at once, regardless of the size as well (Tarique *et al*, 2012).

In Oman, Al-Busaidi and collaborators (2013) tested the presence of metals in clams caught in Sohar, Al Batinah region, which constitutes the major clam fishing area. The authors found maximums of 0.034, 3.92 and 0.13 µg/g wet weight of Hg, Cd and Pb respectively, in the soft tissues of the clam. These assessments were made throughout the year, with higher concentrations for Cd and Pb found during winter months. Levels found for Pb and Hg were within the European standards but levels for Cd were above the limits established for human consumption which makes it mandatory to find the origins of this metal into the marine environment (Al-Busaidi *et al.*, 2013).

A study carried out by Al-Ghassani and collaborators (2013) in a different site of Oman, the Dhofar region, further south close to Yemen, evaluated the concentrations of metals in the rock oyster *Saccostrea cucullata* throughout 1 entire year. The authors found maximum concentrations of 7.08 µg/g, 0.156 µg/g and 0.049 µg/g dry weight of Cd, Pb and Hg, respectively, without significant differences for the season. The maximum concentration of Cd was related to a site where new roads were being built but no other relations were made. Only the level of Cd exceeded the upper level recommended for molluscs and bivalves of 1.0 µg/g wet weight, the other levels were within limits (Al-Ghassani *et al*., 2013).

Another study conducted in the Dhofar region also evaluated the metal concentration in the oyster *Saccostrea cucculata* and found levels for Hg, Cd e Pb, in mean ± sd, of 0.017 ± 0.008 µg/g, 3.31±1.14 µg/g and 0.011 ± 0.03 µg/g, respectively. The highest concentrations of Cd were detected in the months following the monsoon and heavy winds, which lead to higher concentrations of the metal naturally occurring in the environment (Yesudhason *et al.*, 2013). Fowler and collaborators (2007) compared surveys carried out in the Dhofar region with nearly 20 years gap, from 1993 and 2002, to check the trends in heavy metal concentration in seafood, mainly *Sacostrea cucculata* (rock oyster) and *Panulirus homarus* (rock lobster). The authors concluded that concentrations measured during both surveys were for the most part relatively low and did not indicate any major source of contamination. However, the Cd concentrations were consistently high and deserve further control. It is difficult, if not impossible, to establish any consistent temporal trend based on only two surveys spaced 19 years apart (Fowler *et al*., 2007). Nevertheless, these may be useful for assessing any future changes in environmental levels of these contaminants brought about through urban, agricultural and industrial development in the Dhofar region of southern Oman.

A different study conducted between 2007 and 2011 by Al-Mughairi and collaborators (2013) evaluated the concentration of Hg in several shrimps, clams, mussels, oysters and squid from different markets, vending places, supermarkets and factories throughout the country. They found levels in crabs and mussels, in mean ± sd, of 0.27 ± 0.15 and 0.19 ± 0.27 µg/g wet weight respectively, the highest among the other shellfish evaluated (Al-Mughairi *et al.*, 2013).

Nevertheless, all the levels of metal concentrations in the south of Oman are still far below the values found by Mora and collaborators (2004) for the northern site of Oman which can be in part due the north of Oman being a more industrialized area.

All values referred to are expressed in Table 4.2.

Table 2.2 - Mean ± sd or maximum concentrations (µg/g wet weight) found for heavy metals in molluscs, bivalves and crustaceans

Country	**Area**	**Species**	**Metals**				**Reference**	**Exceed limits**
			Pb	**Cd**	**Hg**	**As**		
UAE	Jebel Ali	*Pinna muricata**	1.23	10.7	0.20	153	Mora *et al.*, 2004	Cd, As
	Abu Dhabi	*Pintada radiate**	2.29	2.73	0.09	30.6	Mora *et al.*, 2004	As
Kuwait	Kuwait Bay	*Amiantis umbonella*	45.3± 2.5 (1)	34.7±1.7	1.5 ± 0.9		Tarique *et al.*, 2012	Pb, Cd, Hg
			163.1±4.2 (2)	107.1±1.8	11 ± 1.2			
Saudi	Tarut Bay	*Meretrix meretrix*	2.49±0.27	0.90±0.03			Alyahya *et al.*, 2011	
Arabia	Sharq-dareen	*Penaeus semisulcatus*			0.039±0.012		Al-Saleh & Al-Doush, 2002	
Oman	Ras Al Yei	*Saccostrea cucullata**	0.384	21.9	0.05	17.2	Mora *et al.*, 2004	Cd, As
	Al Sawadi	*Saccostrea cucullata**	0.673	19.9	0.14	11.1	Mora *et al.*, 2004	Cd, As
	Al Batinah	*Liochoncha ornata*	0.13	3.92	0.034		Al-Busaidi, *et al.*, 2013)	Cd
	Dhofar	*Sacostrea cucculata**	0.21	21.2			Fowler *et al.*, 2007	Cd
		*Panulirus homarus**	0.51	0.03				
	Dhofar	*Sacostrea cucculata**	0.156	7.08	0.049		Al-Ghassani *et al.*, 2013	Cd
	Dhofar	*Sacostrea cucculata*	0.011± 0.03	3.31±1.14	0.017±0.008		Yesudhason, *et al.*, 2013	Cd
	All Oman	*Portunus pelagicus*			0.27 ± 0.15		Al-Mughairi, *et al.*, 2013	
		Perna viridis			0.19 ± 0.27		Al-Mughairi, *et al.*, 2013	
Bahrain	BAPCO	*Pintada radiate**	3.92	3.68	0.11		Mora *et al.*, 2004	
		Turbo coronatus	3.03				Al-Sayed & Dairi, 2006	Pb
		Portunus pelagicus[3]	0.035±0.0175	0.0575±0.0315	0.035±0.0175		Musaiger & Al-Rumaidh, 2005	
Qatar	Ras al Nouf	*Circentia callipyga**	1.45	1.17	0.315	156	Mora *et al.*, 2004	Cd, As

* per dry weight; (1)– reference site; (2) - contaminated site; 3 – mg/ 100 g

4.3 Fish

Fish is one of the highest income resources for the population of the GCC, not only in the more industrialized coastal areas but also in less industrialized ones, where fish is nearly the only source of protein for whole families (Al-Yousuf *et al*., 2000). Since fish is on top of the food chain, its heavy metal concentration can be high due to the effect of bio accumulation affecting human health; therefore it is important to monitor its contamination (Zhou *et al*., 2008). Its concentrations depend on the source of contamination and the ability of fish to accumulate such compounds (Bolana *et al.*, 2014).

The contamination of fish with heavy metals, even within levels of concentrations acceptable by the EC, EFSA or even FAO/WHO could become not so acceptable if the amount of fish consumed would substantially increase the exposure to such contaminants. Since fish muscle is the most important part for human consumption and even though organs such as liver may be good indicators of bio accumulation but have little influence on the total body burden (Kosanovic *et al.*, 2007) I will only consider metal concentrations obtained from fish muscle and whole fish samples.

In the year 2000, Al-Yousuf and collaborators tested the presence of Cd, amongst others, in the liver, skin and muscle of *Lethrinus lentjan* fish along the coast of the United Arab Emirates, which extends for 700 km. This fish, commonly called Red Spot Emperor, is one of the most popular fish in the Gulf countries with commercial importance for over 75% of the population (Al-Yousuf *et al*., 2000). The authors found concentrations of 0.11±0.02 µg/g wet weight of Cd in the muscle of the fish and concluded that its concentration had an inverse relation with fish size. In 2004, Mora and collaborators tested fish in two different areas off the coast of the UAE, Al Marfa and Dhannah, among other sites in the Gulf mentioned further. Fish tested are also fish with great commercial value, the orange spotted grouper (OSG), locally known as hamoor, and the spangled emperor (SE), locally known as sharry[2] (Mora *et al.*, 2004). Out of 16 different sites along the Gulf and Gulf of Oman where the authors caught and tested the OSG, Al Marfa had the highest concentration of Hg, 2.35 µg/g dry weight, about 5 times higher than the Hg concentration in the SE in the same place. Dhannah comes in 2nd in the level of Hg in the OSG, 1.62 µg/g dry weight, more than 3 times higher the Hg levels in the SE.

In 2007, Kosanovic and collaborators also tested heavy metal concentration in the Red Spot Emperor along the coast of the UAE. In this study, fish was caught in 3 different sites along the coast and tested for Cd, Pb, Hg and As. The authors found Umm Al Quein to be the area with higher concentrations for Pb and Cd, the last one about two times higher than the values detected by Al-Yousuf and collaborators (2000). Arsenic was found in concentrations of 1.54± 0.54 µg/g wet weight in Dubai area (Kosanovic *et al.*, 2007).

In Kuwait, Al-Majed and Preston (2000) evaluated total and metil-Hg in seven species of fish purchased from local fish markets. Of all species tested, hamoor (*Epinephelus coioides*)

[2] Hamoor and shary are two well-known fish that are always present in the supermarkets and restaurants.

exhibited the highest concentration of all, 3.92 µg/g dry weight of total-Hg and 3.27 µg/g of metil-Hg (Al-Majed & Preston, 2000a).

In 2002, Al-Saleh and Al-Doush tested fish for Hg in three different locations in the eastern coast of Saudi Arabia and found the highest concentration in *Acanthoparagus bifasciatus* (doublebar-bream) with 0.198±0.011 µg/g wet weight, mean ±sd, in Sharq-dareen followed by Mounifah, with 0.111±0.013 µg/g wet weight in *Epinephelus tauvina* (hamoor), also one of the most popular and economically important in Saudi Arabia, both below the maximum permitted levels established by authorities (Al-Saleh & Al-Doush, 2002).

Al-Saleh and Shinwari (2002) also tested the *Epinephelus tauvina*, as well as *Lethrinus miniatus, Siganus canaliculatus* and *Acanthoparagus bifasciatus*, all caught from Maniefa (= Mounifah), Dareen and Al-Dammam. The authors found concentrations, in mean ± sd levels, of Cd, Pb and As of 4.6±7.2, 20.0±20.8, and 42.7±17.4 ng/g wet weight respectively, in all fish samples (Al-Saleh & Shinwari, 2002). The results of this method showed that the concentrations of Cd, Pb and As were significantly higher in Maniefa than in the other two sites which confirms that Maniefa site is affected by industrial waste discharge taking place in that area. Nevertheless the values are presented in nanograms, which translates to a very low number in micrograms (1 ng= 0.001 µg).

In 2006, Ashraf tested heavy metal concentration in tuna fish caught by commercial vessels from the coasts of KSA destined to be canned at a commercial factory on land. The author found levels of Pb, Cd and Hg of 0.53±0.08, 0.16±0.11 and 0.31±0.17 µg/g in mean ± sd, respectively (Ashraf, 2006).

In 2010, Al Othman checked some foods from different markets in Riyadh City for Pb contamination. The author found mean ± sd levels of Pb in fresh fish of 0.068 ± 0.051 µg/g dry weight. The author also tested 6 different brands of sardines and tuna caught and canned in the KSA and found the highest level of Pb in sardines, with 0.063±0.015 µg/g dry weight (Al-Othman, 2010).

In the most recent paper on heavy metal concentration in fish from Saudi Arabia, Mahboob (2014) evaluates the concentration of Pb, Cd, Hg and As in the muscle of *Oreochromis niloticus* (tilapia), *Clarias gariepinus* (catfish), *Poecilia latipinna* (sailfin molly) and *Aphanius dispar dispar* (killifish), all freshwater fish. These were caught in Wadi Hanifah[3], a natural watershed with approximately 4,400 km^2 that goes right through the city of Riyadh, in both pre and post-monsoon periods. Average levels of heavy metals found ranged from 0.427 µg/g, 0.760 µg/g and 1.125 µg/g wet weights of Cd, Pb and As, respectively (Mahboob *et al*, 2014). Results showed no significant impact of the period of the year on the levels of contamination of the fish. *P. latipinna* and *A. dispar dispar* show highest levels of Cd and Pb, though these are not commercially important due to the eating preferences of local population, despite being amongst the most abundant freshwater fish species in the country (Mahboob *et al.*, 2014).

[3] A Wadi is a valley or streambed that remains dry except during the rainy season. When it finds a contained area, if it is deep and the soil made of hard rock, the water will not be absorbed and it will form a dam which then becomes the habitat for several species.

Fish from Oman was also part of the big study from Mora and collaborators (2004). The authors tested *Epinephelus coioides* (hamoor) and *Lethrinus nebulosus* (shary) from four different places including one Port. Raysut Port Area only showed the highest level for Hg, 0.522 µg/g dry weight and Sagar area showed the highest levels of As and Cd, 2.88 and 0.014 µg/g dry weight, respectively (Mora *et al*., 2004).

Later in 2011, Al Busaidi and collaborators tested the muscle of fresh and frozen fish samples from seafood processing units in Oman, all with commercial importance, including fish imported from different countries. The authors tested skipjack tuna (*Katsuwonus pelamis*), yellowfin tuna (*Thunnus albacares*), red seabream (*Pagellus affinis*), santerbream (*Cheimerius nufar*), grouper (*Epinephalus chlorostigma*), emperor (*Lethrinus nebulosus*), amberjack (*Seriola dumerili*), catfish (*Arius bilineatus*), yellowtail scad (*Atule mate*) and kingfish (*Scomberomorus commerson*) (Al-Busaidi *et al.*, 2011). Skipjack tuna showed lower Cd concentrations with a mean concentration of 0.0049 ±0.010 µg/g wet weight. The highest concentration was detected in yellow fin tuna with 1.0 µg/g which can be due to their feeding habits, as big species feed at higher tropic levels and is expected that they accumulate higher levels of bioaccumulative metals. Mean Cd concentration is between 0.0049±0.010 and 0.0364±0.0001 µg/g wet weight. Regarding Pb among individual species, yellowfin tuna contained the lowest Pb concentration with mean tissue concentration of 0.0291± 0.127 µg/g wet weight. Santer bream and emperor both contained the highest mean Pb concentration of 0.196 ± 0.310 µg/g and 0.180 ± 0.190 µg/g, respectively. The highest Pb concentration was found in skipjack tuna at a level of 1.437 µg/g and the lowest Pb concentration was found in amberjack with 0.083 µg/g wet weight. The highest Hg concentration was detected in amberjack with a mean value of 0.101± 0.167 µg/g and the lowest with grouper, 0.015±0.053 µg/g (Al-Busaidi *et al.*, 2011).

Al Mughairi and collaborators (2013) also tested commercial fish and seafood for Hg. The authors tested 31 different species for fish and seafood collected from fish-landing centres, seafood factories, canning factories, dry-fish markets and supermarkets. Of fresh fish tested the authors found levels of 0.07±0.10 µg/g wet weight Hg, in mean ± sd. None of the fresh, frozen, dried or canned fish has higher values than the ones defined by authorities (Al-Mughairi *et al.*, 2013).

More recently, in 2013, Abdul-Wahab and collaborators investigated metal contamination in muscle tissue of the grouper *Epinephelus coioides* in the vicinity of a single buoy mooring (SBM3) at the Sultanate of Oman. Mina Al Fahal is of primary importance to the economy of Oman because it works as an export point of crude oil via terminal and offshore operations at single buoy moorings and is the site of Petroleum Development Oman's oil storage facility (tank farm), tanker loading operations and the Oman Refinery Company. Hence, the risk of its metal contamination is an expected issue. The grouper is one of the most commonly consumed fish in the Sultanate. Mina Al Fahal is an open sandy bay within the capital area, Muscat, which also serves as fishing area. The authors found mean (±sd) concentrations of Cd and Pb of 0.05±0.004 and 0.20±0.018 µg/g dry weight, respectively (Abdul-Wahab *et al*., 2013).

The chemical composition of fish was also studied in Bahrain. Musaiger and D'Souza (2008) tested fish bought in Manama City from a local market. Only muscle tissue was analysed. The author found that most fish had low levels of heavy metals, with maximums of 0.03 µg/g wet weight for Cd found on *Rhabdsargus haffara* (locally knowns as Qurqufan), 0.20 µg/g wet weight of Hg found in *Epinephelus areolatus* (knowns as hammour) and 0.50 µg/g wet weight Pb found in *Liza alata* (locally knowns as Maid) (Musaiger & D'Souza, 2008). All these fish are commonly consumed in Bahrain. This study concludes that all levels are within range and accordingly to levels found by Ashraf (Ashraf, 2005).

It is important to point out that Musaiger and D'Souza (2008) refer to the values found in muscle tissue when Sharaf evaluated levels found in kidneys and heart tissue. When these numbers are compared to the levels found by Mora and collaborators (2004) one might think that either the sea has become extremely clean or there might be some limitations with Musaiger and D´Souza´s results. Mora and collaborators (2004) found high levels of Hg in the muscle of the *Epinephelus coioides* (different species but same genus, also commonly known as hammoor) between 0.669 and 0.820 µg/g dry weight, for fish caught in Fasht Al Adham and Badaiya, respectively (Mora *et al.*,2004). Badaiya is located on the western coast of Bahrain, is less populated than Manamah City, which eventually would be less contaminated. Fasht al Adham is located on the eastern coast, farther south and again is less populated. So either the fish used by Musaiger and D´Souza came from an uncontaminated site or the rehearsal was badly conducted. Other levels found by Mora and collaborators (2004) in Bahrain area, found levels of Pb and Cd within range but found levels of As of 14.4 µg/g dry weight in Fashat AL Dahm area (Musaiger & Al-Rumaidh, 2005).

In 2001, several species of fish were bought in local fish markets in Qatar. Al-Jedah and Robinson found very satisfactory levels of mercury in all fish, the highest value found in the muscle of hamoor (*Epinephelus tauvina*) from Doha, 0.24 µg/g, well below the limits defined by authorities (Al-Jedah & Robinson, 2001). In 2004, when Mora and collaborators tested the hamoor and sharry, they found that hamoor in the Al Khawr area had a concentration of Hg of 1.04 µg/g dry weight, the highest for Qatar places that were tested. A high level of As was found in sharry from Al Dakhira, 10 µg/g dry weight. The authors conclude that As values for this area are among the highest from literature, but as previously reported, there is very limited data on As values, which is clearly a topic that deserves further investigation (Mora *et al.*, 2004).

Heavy metal concentration in fish muscle and respective comparison with maximum permitted levels can be found in Table 4.3.

Table 4.3 - Mean (± sd) or maximum heavy metal concentration (µg/g wet weight) found in fish muscle along the coast of the Gulf and Gulf of Oman

Country	**Area**	**Species**	**Metal**				**Reference**	**Exceed limits**
			Pb	**Cd**	**Hg**	**As**		
UAE	Ras Al Khaima	*Lethrinus lentjan*		0.11±0.02			Al-Yousuf *et al.*, 2000	Cd
	AL Marfa	*Epinephelus coioides**	0.025	<0.001	2.35	4.1	Mora *et al.*, 2004	
	Dhannah	*Epinephelus coioides**		<0.001	1.62	1.9	Mora *et al.*, 2004	
	Dubai	*Lethrinus lentjan*	0.004±0.002	0.17±0.06	0.05±0.012	1.54± 0.54	Kosanovic *et al.*, 2007	Cd
	Sharjah	*Lethrinus lentjan*	0.005±0.004	0.13±0.05	0.068±0.035	0.839±0.436	Kosanovic *et al.*, 2007	Cd
	Umm Al Quein	*Lethrinus lentjan*	0.018±0.011	0.19±0.05	0.036±0.025	1.28±0.122	Kosanovic *et al.*, 2007	Cd
Kuwait	Kuwait Bay	*Epinephelus coioides**			3.92		Al-Majed & Preston, 2000a	Hg
Saudi Arabia	Mounifah	*Epinephelus tauvinaa*			0.111±0.013		Al-Saleh & Al-Doush, 2002	
	Sharq-dareen	*Acanthoparagus bifasciatusa*			0.198±0.011		Al-Saleh & Al-Doush, 2002	
	Maniefa	All tested species b	20.0±20.8	4.6±7.2		42.7±17.4	Al-Saleh & Shinwari, 2002	
	Coasts of KSA	Tuna fish	0.53±0.08	0.16±0.11	0.31±0.17		Ashraf, 2006	Pb, Cd
	Riyadh City	Fresh fish *	0.068±0.051				Al-Othman, 2010	
		Sardines	0.063±0.015					

Table 4.3 (continued) - Mean (± sd) or maximum heavy metal concentration (µg/g wet weight) found in fish muscle along the coast of the Gulf and Gulf of Oman

Country	Area	Species	Metal				Reference	Exceed limits
			Pb	Cd	Hg	As		
	Wadi Hanifah	Several	0.760	0.427		1.125	Mahboob *et al.*, 2014	Cd
Oman	Raysut Port	*Lethrinus nebulosus**	0.014	0.011	0.522	2.07	Mora *et al.*, 2004	
	Sagar		0.011	0.014	0.435	2.88		
	Processing units	*Scomberomorus commerson*	0.1557±0.156	0.0364±0.001	0.0677±0.03		Al-Busaidi *et al.*, 2011	
		Cheimerius nufar	0.1964±0.310	0.0099±0.014	0.0926±0.022			
		Seriola dumerili	0.068±0.042	0.0122±0.020	0.1016±0.167			
	Mina Al Fahal	*Epinephelus coioides**	0.20±0.018	0.05±0.004			Abdul-Wahab *et al.*, 2013	
	All Oman	Fresh fish			0.07±0.10		Al-Mughairi *et al.*, 2013	
		Canned fish			0.07±0.08			
		Dried fish*			0.23±0.60			
Bahrain	Fasht Al Dahm	*Epinephelus coioides**	0.028	0.001	0.669	14.4	Mora *et al.*, 2004	As
	Badaiya	*Epinephelus coioides**	0.005	0.001	0.820	1.38	Mora *et al.*, 2004	
	Manama City	*Liza alata*	0.50	0.02	0.02		Musaiger & D´Souza, 2008	
		Epinephelus areolatus	0.02	0.02	0.20			
		Rhabdosargus haffara	0.02	0.03	0.03			

Table 4.3 (continued) - Mean (± sd) or maximum heavy metal concentration (µg/g wet weight) found in fish muscle along the coast of the Gulf and Gulf of Oman

Country	**Area**	**Species**	**Metal**				**Reference**	**Exceed limits**
			Pb	**Cd**	**Hg**	**As**		
Qatar	Doha	*Epinephelus tauvina*			0.24		Al-Jedah & Robinson, 2001	
	Al Khwar	*Epinephelus coioides**	0.113	0.013	1.04	3.1	Mora *et al.*, 2004	
	Al Dakhira	*Lethrinus nebulosus**	0.108	0.005	0.343	10.0	Mora *et al.*, 2004	As

* per dry weight; a – sample taken from whole fish tissue; b – ng/g wet weight, whole tissue

5. ESTIMATED EXPOSURE ASSESSMENT FOR HUMAN CONSUMERS

In general, risk assessment of dietary fish and metal intake are examined with prolonged exposure and not based on one average meal. However, there is an increasing concern that one average meal of fish and seafood, with a pulsed exposure, could severely affect our health. As mentioned before, higher and lower levels of metals are found in different species from different areas along the Gulf and the Gulf of Oman into the Arabian Sea. From the public health point of view, heavy metal concentrations in many of the analysed samples of seafood were well within the prescribed limits set by various authorities, others were not.

Regarding surveys on metal quantification in humans in these countries there aren´t many studies available. In some articles, it is not specified the way of intake, if oral, respiratory or other, neither the contribution of fish to that intake. Food frequency questionnaires are scarce and, as these countries have enormous numbers of expatriates, it is difficult to determine if it represents only GCC nationals. These surveys should also assess oral intakes of different ages and genders. Some studies have assessed the concentration of metals in breast milk, umbilical cord and placenta of women, as well as in their infants. For mercury, levels in toenails or hair provide the best biomarkers of chronic mercury exposure, given their slow growth (Mozaffarian & Rimm, 2006). Some studies have evaluated concentration in hair of children and adults. Only a few have included diet as a variable to relate to.

An assessment of Hg in 100 hair samples from fishermen in Kuwait carried out in the year 2000 by Al-Majed and Preston found that 78% of them had Hg levels around twice the reference mean (Al-Majed & Preston, 2000b). These fishermen were assessed for the number of meals that consisted of fish; to be noted that they ate from 1 meal of fish per day to a maximum of 3 meals of fish per day. This is not the case for the general population, but it is probably for the majority of fishermen throughout the GCC.

In 2003, Al-Saleh and collaborators determined Pb, Cd and Hg concentrations in breast milk of Saudi lactacting mothers and their babies, from Riyadh and Al-Ehssa regions (Al-Saleh *et al*., 2003). The estimated weekly intake of metals of breast fed babies were in some cases higher than the PTWI recommended, which pose a threat to their health due to the rapid growth, immaturity of their organs and the susceptibility of their nervous system. Among the independent variables included in this study, the only ones with significant influence on Cd, Pb and Hg concentrations were the region and the consumption of fish. The limitation of this survey is that it doesn’t have any data regarding the consumption frequency of fish making it impossible to establish a pattern. Regardless of the lack of information about the dietary intake, but considering the results, the authors consider infants and lactating women within the risk groups and call for preventive measures where heavy metals could be identified and eliminated (Al-Saleh *et al.*, 2003).

In 2005/2006, Al-Saleh and collaborators determined the concentration of metals in maternal blood, cord blood and placenta of healthy women admitted for delivery in a central hospital in Saudi Arabia (Al-Saleh *et al*., 2011). The authors questioned the participants on the consumption of seafood (responded as Yes or No) and found significant influence on the maternal blood Cd and Hg concentrations and as well in placental Hg levels. Also placental Pb levels were positively associated with maternal Hg levels. Cord blood Pb levels have been used as an index of prenatal Pb exposure. Numbers reported in this study are less than half comparing to reported cord blood and maternal blood Pb levels in the vicinity of a solid waste incinerator in Portugal, only exceeded by Karachi, Pakistan (Reis *et al.*, 2007; Al-Saleh *et al*., 2011). Regarding Cd, the authors refer that since none of the mothers were occupationally exposed to it, their exposure might have come from either dietary or environmental sources. Last, elevated levels of Hg are explained by the high consumption of fish, since levels were higher than in the non-fish eaters. Results in this study showed different predictors for heavy metal exposure. Maternal Hg was a significant predictor of placental Pb, suggesting that either one is influenced by the other or the two share the same source of exposure. This brings awareness about the potential risk from exposure to a mixture of metals and their toxicological interactions. Eating seafood was among the predictors for maternal Cd exposure, which in turn was one of the predictors, along with placental Pb and Hg, for placental Cd levels. This study suggested that the variability found could come from sources not taken into account and factors like diet should be explored since they contribute to maternal and fetal exposure (Al-Saleh *et al.*, 2011).

Between 2011 and 2012, Al-Saleh and collaborators evaluated Hg in breast milk and urine of health women and respective infants. More than 57% of women had milk with concentrations above the background level of Hg in human milk and 4.5% of the infants exceeded the limit for consumption. The majority of lactating mothers in this study consumed fish but the frequency in most of them was “irregular” or “once a month”. The ratio for breast milk/ blood is greater than 1 which indicates that levels of Hg are higher in milk than in blood suggesting a transfer of the Hg from blood to milk increasing the exposure to infants which may be responsible for many health problems especially during the neurodevelopment period (Al-Saleh *et al*., 2013).

Foodborne exposure to MeHg was also determined in the UAE (Davidson *et al*., 2012) by analysing fruits, vegetables, fish and seafood consumption. Regarding the UAE there are no data available that describe the incidence of contamination of foods. This study conducted a survey to perceive the consumption frequency and combined nutrition data for daily portion sizes and age related modifying coefficient was used to account for age related differences in the amount of food consumed. Exposure risk varied with age, gender and weight. This survey suggests that of the total population enquired, 69% male and 31% women could be at risk of overexposure to methyl-Hg from eating seafood. This study does not take into account the benefits of the consumption of seafood that may outweigh the risks and assumes that all UAE residents have similar habits to the rest of the Emiratis, which is not true, due to the constant renew of the population and the mixed origins present and as well because interior populations still rely on camel milk, wheat and dates (Davidson *et al.*, 2012).

Some studies, that evaluate heavy metal concentration in human samples such as hair, demonstrated that there are alterations in toxic heavy metals and essential trace elements in autistic children's hair from Saudi Arabia and Oman (Al-Ayadhi, 2005; Al-Farsi *et al.*, 2013). Another detects significant proportion of pregnant women with Pb levels above safety limits in Kuwait (Rahman *et al*., 2012), another finds healthy women in Saudi Arabia with Hg levels above safety limits (Al-Saleh *et al*., 2006) and another finds Pb in school girls (Al-Saleh *et al*., 2001).

To assess public health impact of metals in fish, it is essential to estimate the daily intake (Estimated Daily Intake, EDI) of metals by humans expressed as µg/day. This can be obtained by multiplying the average quantity of fish consumed per capita per day by the concentration of metal. That quantity is then calculated per week (Estimated Weekly Intake, EWI) to compare with the PTWI for each metal. Concentrations per dry weight were previously converted into wet weight concentrations. All EWIs were obtained using the apparent consumption of fish and seafood for each country reported by FAO statistics. For the comparison of the EWI per capita with the PTWI for each metal, it was assumed by default a person with 60 kg (JECFA, 2011).

The EWIs for molluscs, bivalves, crustaceans and fish are presented in Table 5.1 and Table 5.2, respectively.

Table 5.1 - EWI of Pb, Cd, Hg and As from molluscs, crustaceans and bivalves in µg/ per capita from mean concentrations (µg/g wet weight) of metals present in tested samples

Reference	Country	Area	Species	Pb		Cd		Hg		As	
				[Mean]	**EWI**	**[Mean]**	**EWI**	**[Mean]**	**EWI**	**[Mean]**	**EWI**
Mora *et al.*, 2004	UAE	Jebel Ali	*Pinna muricata*	0,256	140,553	2,229	1222,698	0,042	22,854	31,875	17483,438
Mora *et al.*, 2004		Abu Dhabi	*Pintada radiate*	0,477	261,680	0,569	311,959	0,019	10,284	6,375	3496,688
Tarique *et al.*, 2012	Kuwait	Kuwait Bay	*Amiantis umbonella*	(1) 45,300	15551,490	34,700	11912,510	1,500	514,950		
				(2) 163,100	55992,230	107,100	36767,430	11,000	3776,300		
Alyahya, *et al.*, 2011	Saudi Arabia	Tarut Bay	*Meretrix meretrix*	0,519	81,548	0,900	141,480				
Al-Saleh & Al-Doush, 2002		Sharq-dareen	*Penaeus semisulcatus*					0,039	6,131		
Mora *et al.*, 2004	Oman	Ras Al Yei	*Saccostrea cucullata*	0,080	44,032	4,563	2511,200	0,010	5,733	3,583	1972,267
Mora *et al.*, 2004		Al Sawadi	*Saccostrea cucullata*	0,140	77,171	4,146	2281,867	0,029	16,053	2,313	1272,800
Al-Busaidi, *et al.,* 2013		Al Batinah	*Liochoncha ornata*	0,130	71,552	3,920	2157,568	0,034	18,714		

Table 5.1 (continued) - EWI of Pb, Cd, Hg and As from molluscs, crustaceans and bivalves in µg/ per capita from mean concentrations (µg/g wet weight) of metals present in tested samples

Reference	Country	Area	Species	Pb		Cd		Hg		As	
				[Mean]	**EWI**	**[Mean]**	**EWI**	**[Mean]**	**EWI**	**[Mean]**	**EWI**
Fowler *et al.,* 2007	Oman	Dhofar	*Sacostrea cucculata*	0,044	24,080	4,417	2430,933				
			Panulirus homarus	0,106	58,480	0,006	3,440				
Al-Ghassani *et al.*, 2013		Dhofar	*Sacostrea cucculata*	0,033	17,888	1,475	811,840	0,010	5,619		
Yesudhason, *et al.*, 2013		Dhofar	*Sacostrea cucculata*	0,110	60,544	3,310	1821,824	0,017	9,357		
Al-Mughairi, *et al.*, 2013		All Oman	*Portunus pelagicus*					0,270	148,608		
Al-Mughairi, *et al.*, 2013			*Perna viridis*					0,190	104,576		
Mora *et al.*, 2004	Bahrain	BAPCO	*Pintada radiate*	0,817	211,517	0,767	198,567	0,023	5,935		
Al-Sayed & Dairi, 2006			*Turbo coronatus*	3,030	784,770						
Musaiger & Al-Rumaidh, 2005			*Portunus pelagicus*	0,350	90,650	0,575	148,925	0,350	90,650		

Table 5.1 (continued) - EWI of Pb, Cd, Hg and As from molluscs, crustaceans and bivalves in µg/ per capita from mean concentrations (µg/g wet weight) of metals present in tested samples

Reference	Country	Area	Species	Pb		Cd		Hg		As	
				[Mean]	EWI	[Mean]	EWI	[Mean]	EWI	[Mean]	EWI
Mora *et al.*, 2004	Qatar	Ras al Nouf	*Circentia callipyga*	1,450	681,210	1,170	549,666	0,315	147,987	156,0	73288,8

(1) – Reference site; (2) – contaminated site

Table 5.2 - EWI of Pb, Cd, Hg and As from fish muscle in µg/ per capita from mean concentrations (µg/g wet weight) of metals present in tested samples

Reference	Country	Area	Species	Pb		Cd		Hg		As	
				[Mean]	EWI	[Mean]	EWI	[Mean]	EWI	[Mean]	EWI
Al-Yousuf *et al.*, 2000	UAE	Ras Al Khaima	*Lethrinus lentjan*			0,110	60,335				
Mora *et al.*, 2004		AL Marfa	*Epinephelus coioides*	0,005	2,86	0,000	0,114	0,490	268,536	0,854	468,510
Mora *et al.*, 2004		Dhannah	*Epinephelus coioides*			0,000	0,114	0,338	185,119	0,396	217,115
Kosanovic *et al.*, 2007		Dubai	*Lethrinus lentjan*	0,004	2,19	0,170	93,245	0,050	27,425	1,540	844,690
Kosanovic *et al.*, 2007		Sharjah	*Lethrinus lentjan*	0,005	2,74	0,130	71,305	0,068	37,298	0,839	460,192
Kosanovic *et al.,* 2007		Umm Al Quein	*Lethrinus lentjan*	0,018	9,87	0,190	104,215	0,036	19,746	1,280	702,080
Al-Majed & Preston, 2000a	Kuwait	Kuwait Bay	*Epinephelus coioides*					0,817	280,362		
Al-Saleh & Al-Doush, 2002	Saudi Arabia	Mounifah	*Epinephelus tauvinaa*					0,111	17,449		
Al-Saleh & Al-Doush, 2002		Sharq-dareen	*Acanthoparagus bifasciatusa*					0,111	17,449		
Al-Saleh & Shinwari, 2002		Maniefa	All tested species	0,004	0,66	0,001	0,151		0,000	0,043	6,712
Ashraf, 2006		Coasts of KSA	Tuna fish	0,530	83,32	0,160	25,152	0,310	48,732		

Table 5.2 (continued) - EWI of Pb, Cd, Hg and As from fish muscle in µg/ per capita from mean concentrations (µg/g wet weight) of metals present in tested samples

Reference	Country	Area	Species	Pb		Cd		Hg		As	
				[Mean]	EWI	[Mean]	EWI	[Mean]	EWI	[Mean]	EWI
Othman, 2010			Sardines	0,063	9,90						
(Mahboob *et al.*, 2014)		Wadi Hanifah	Several	0.760	119.472	0.427	13.984			1.125	176.85
Mora *et al.*, 2004	Oman	Raysut Port Area	*Lethrinus nebulosus*	0,003	1,61	0,002	1,261	0,109	59,856	0,431	237,360
Mora *et al.*, 2004		Sagar	*Lethrinus nebulosus*	0,002	1,26	0,003	1,605	0,091	49,880	0,600	330,240
Al-Busaidi *et al.*, 2011			*Scomberomorus commerson*	0,156	85,70	0,036	20,035	0,068	37,262		
Al-Busaidi *et al.*, 2011			*Cheimerius nufar*	0,196	108,10	0,010	5,449	0,093	50,967		
Al-Busaidi *et al.*, 2011			*Seriola dumerili*		0,00	0,012	6,715	0,102	55,921		
Abdul-Wahab *et al.*, 2013		Mina Al Fahal	*Epinephelus coioides*	0,042	22,93	0,010	5,733				
Al-Mughairi *et al.*, 2013		All Oman	Fresh fish					0,070	38,528		
			Canned fish					0,070	38,528		
			Dried fish					0,048	26,373		
Mora *et al.*, 2004	Bahrain	Fasht Al Dahm	*Epinephelus coioides*	0,006	1,51	0,000	0,054	0,139	36,098	3,000	777,000
Mora *et al.*, 2004		Badaiya	*Epinephelus coioides*	0,001	0,27	0,000	0,054	0,171	44,246	0,288	74,463

Table 5.2 (continued) - EWI of Pb, Cd, Hg and As from fish muscle in µg/ per capita from mean concentrations (µg/g wet weight) of metals present in tested samples

Reference	Country	Area	Species	Pb		Cd		Hg		As	
				[Mean]	EWI	[Mean]	EWI	[Mean]	EWI	[Mean]	EWI
Al-Jedah & Robinson, 2001	Qatar	Doha	*Epinephelus tauvina*					0,240	112.752		
Mora *et al.*, 2004		Al Khwar	*Epinephelus coioides*	0,024	11,06	0,003	1,272	0,217	101,790	0,646	303,413
Mora *et al.*, 2004		Al Dakhira	*Lethrinus nebulosus*	0,025	11.55	0.001	0.489	0.071	33.571	2.083	978.750

6. DISCUSSION

Assuming that molluscs have approximately the same moisture content as fish, around 80% and as some of the concentrations are expressed as µg/g dry weight, in order to evaluate if they are above or below the permitted levels for metal concentration in fish and seafood defined by authorities, the concentrations expressed as µg/g dry weight had to be converted to concentrations as µg/g wet weight, using the conversion factor 4.8 according to Rahman and collaborators (2012) (Rahman *et al*., 2012).

After comparing all concentrations with the maximum permitted levels for molluscs and crustaceans, I find that all samples that were tested for As showed concentrations above the maximum permitted levels of 2.0 mg/ kg wet weight, the most contaminated sites being Ras al Nouf in Qatar and Jebel Ali in the UAE. Whether or not the As present is due to the geological characteristics of the area, the fact is that seafood is contaminated and we ingest it. Aside from the samples expressed here, no other data is available for As in bivalves and crustaceans, so it would be worthwhile to continue to examine its concentration in these and other species, given the hazard effects of this metal in health. All samples caught from Kuwait bay, were highly contaminated with Pb, Cd and Hg, with significant difference between the reference site and the contaminated site. Kuwait bay is a semi enclosed area which extends for 70 km and it is characterized by mudflats specifically in the shallow northern part, which is less than 5 meters deep. Most studies conducted on the coast of Oman showed concentrations of Cd above the maximum permitted levels as did the sample from Qatar and Jebel Ali in the UAE. The concentrations found exceeded the threshold values for Cd in fish and seafood, 1.0 µg/g wet weight established by the EC (Comissão Europeia, 2006). For Pb, apart from Kuwait, only Bahrain samples of *Turbo coronatus* were above the maximum permitted levels (Table 4 and Table 6).

Assuming the apparent consumption of fish and seafood, including fish, molluscs, crustaceans and bivalves for each member of the GCC, the PTWI for each metal and the samples tested, I find that the EWI of these metals from shellfish may largely overcome the PTWI for them and the concern is that it may happen from one meal alone, if it is composed of shellfish. Clearly, those groups that include fish and seafood in nearly all meals are at a greater risk, such as fishermen. Considering the PTWI for a 60Kg person I find that, from the consumption of these samples of shellfish, the EWI for As is largely above the PTWI for all samples. For Cd, all except some from Oman, Bahrain and KSA entail a EWI above the PTWI. None of the samples, except those from Kuwait Bay provide Pb above the PTWI and for Hg, only Venus clams (*Circentia callipyga*), the Blue Crab (*Portunus pelagicus*) and the mussel (*Perna viridis*) contain higher amounts with a EWI above the PTWI.

Regarding fish contamination, after the conversion for a concentration of µg/g wet weight, I can see that despite most locations provide fish with metal concentration within permitted levels, nine locations had fish containing at least one metal in concentrations above maximum permitted levels. Four locations in the UAE contained fish that had Cd above permitted levels, the maximum found in Umm Al Quein, probably because of the characteristics of the fish, since fish of the genus *Lethrinus* are demersal and carnivores. Also tuna fish from KSA and fish from Wadi Hanifah, which only contains fresh water fish, had higher levels of Cd. Tuna caught off the coast of KSA also presented higher levels of Pb. Being tuna a predator, it could be one of the main reasons for such concentration. Highest levels of Hg could only be found in fish caught in Kuwait Bay and as for As, the highest level was found in fish caught in the Fasht Al Dahm area in Bahrain followed by Al Dakhira in Qatar. Fish caught off the coasts of Oman did not present any metals in concentrations above maximum permitted levels. Nevertheless, levels of metals in fish vary due to various factors, including feeding habits, species, age and size of fish, bioavailability of chemicals in food and water (Abdul-Wahab *et al*., 2013). It is expected that fish living higher in the tropic level and bigger in size would accumulate higher levels of metals, like tuna for example (Al-Busaidi *et al.*, 2011) so it could be possible that this fish, if caught off the coast of Oman may present some metals in higher concentrations.

Regarding fish consumption and the weekly intake of heavy metals from fish I find that only the estimated weekly intake of Hg and As is above the recommended PTWI for humans, especially fish from the UAE, Kuwait and Qatar for Hg and fish from UAE and Bahrain for As. Even though the consumption of fish does not reflect an intake of heavy metals above recommended levels, if it is added the consumption of shellfish, these numbers increase even more and the levels ingested may become hazardous. There is also the risk, depending on the amount consumed, that only one meal could provide an intake of metals above recommended.

These EWIs are important for the setting of recommendations for special groups like pregnant women and children living in the GCC. Regarding fish it is not so worrisome since fish consumption also has several benefits for our health. A part from the high biological value protein, results from case-control studies, prospective cohort studies and randomized controlled trials indicate that a modest consumption of fish or fish oil lowers the risk of cardiac mortality. A modest consumption (~250 mg/d) of the marine n-3 PUFA, eicosapentaenoic acid (EPA) and docosahexaenoic acid (DHA) significantly lowers this risk. The quantity of fish servings needed to consume an average of 250 mg/d EPA+DHA varies depending on the particular fish species, but for fatty fish (e.g., anchovies, herring, salmon, sardines, trout, white tuna) is 1-2 servings of 100g/ week (Mozaffarian & Rimm, 2006). The apparent consumption for fish in these countries is well above this dose, except for KSA. Fish and seafood also contain selenium, an essential trace mineral with an effect on reducing cardiovascular disease. This protection may come from the antioxidant effects that could minimize the damage in the cells induced by metals. The intake of selenium and vitamins B decrease the intestinal absorption of As so probably having good nutrition habits could decrease the deleterious effect of the exposure to this metal (Sabath & Robles-Osorio, 2012).

Based on the current evidence, the health risks for adults of not consuming fish outweigh potential risks from metals (Mozaffarian, 2009). Regarding shellfish, given the high concentration of metals I would recommend to maintain the consumption at the lowest level possible and recommend children, pregnant and lactating women to avoid consuming shellfish at all. Also, metal effects are reduced by antioxidants, so it would be important to know the consumption patterns of other food rich in antioxidants to assess the real risk of consumption of food contaminated with metals. If the population is educated into adopting healthy eating habits it may be possible to minimize the hazardous effects of contaminants from food.

There are several limitations that need to be taken into account before any conclusion. While conducting a collection of papers on the contamination of fish and seafood I encountered several inconsistencies. Among the different studies, a diversity of epidemiologic design and analytic methodologies could be observed, making data comparison difficult and not allowing a precise pattern of contamination. Sample preparation steps are usually the most common source of analytic error, which were beyond my control. Regarding studies conducted to evaluate levels of metals in human samples, they were heterogenic in gender, age and socioeconomic status.

The assessment of exposure to food toxicants or contaminants requires data on the dietary intake of food items or groups that are known or are most likely to contain the metal of interest. There are three basic approaches to determining dietary intake: (1) total diet study, (2) survey of individual households or individuals, using prospective food records or dietary recall, and (3) duplicate diet studies. As already mentioned, there aren´t many data available on food frequency questionnaires.

Data on dietary intake needs to be combined with databases (e.g., from governmental monitoring programs) to determine the concentration of the metal in the food. These databases may not be complete or need to be adjusted, for which new surveys need to be conducted. Hence, it is possible that the overall associations might have been overestimated or underestimated. The dietary exposure is calculated by multiplying a fixed value for consumption of a food (usually the mean population value) with a fixed value for the chemical concentration in that food assuming a specific body weight (standard human = 60 kg). As these are such big countries, especially Oman, the UAE and Saudi Arabia, the consumption of fish is very different between coastal and the interior populations. The average apparent consumption is defined for the entire country and may not reflect a real consumption pattern. As well, the body weight defined to calculate the PTWI may not reflect the average body weight of the majority of the population given the rise in obesity numbers in the last years. Once dietary exposure data are available, the next step is to determine whether this level of exposure constitutes a human health risk. For many food toxicants and contaminants, data on their toxicities are only available from studies in laboratory animals, most commonly rodents. Further, publications may have been missed.

To increase the accuracy, consistency, and reproducibility of future studies, I would recommend that collection protocols and analytical methods for metal assessment in human samples become harmonized to provide a scientific basis for inter-laboratory comparisons and to improve risk assessment strategies to address different exposure scenarios.

7. CONCLUSION

Heavy metal levels in various types of living organisms from the waters surrounding the GCC have been investigated by several researchers. Some studies indicated that both in algal species and sediments heavy metals are present in low concentrations. However, higher levels of heavy metals have been detected in localized areas influenced by industrial facilities, desalination plants and oil refineries. Other studies detected a trend of increasing metal concentrations, suggesting evidence of contamination from anthropogenic sources.

Most of the relevant studies conclude that heavy metals in fish tissues were within allowable concentrations and pose no threat to public health. However, some studies reported levels of metals that exceeded the permissible limits. Hotspots of heavy metal contamination were identified in localized areas influenced by oil pollution from refiners and intensive dredging or recreation activities. Therefore, regular monitoring of heavy metal levels in fish species seems necessary to prevent health risks and to ensure nutritional safety conditions. Overall, cumulative information available on the levels of heavy metals in living organisms and sediments is generally irregular and does not provide a complete picture of their spatial and temporal distribution in the waters of Gulf, Gulf of Oman and the Arabian Sea. Hence, there is a critical need for an approach to monitor heavy metal concentrations and distributions. Given the little information about heavy metal load amongst GCC nationals, there is also a critical need to identify the major consumption patterns of the population in these countries.

These countries are under great industrial transformation. Fish and seafood apparent consumption may not be as described and may change very fast. As wealth increases food patterns may change. The typical diet becomes diversified and so does the production. Until recently food patterns among coastal populations was basically fish and into the interior and desert it was based on wheat, dates and camel protein. These changes may increase the potential risk for exposure to metals in food. It would be recommended to conduct food frequency questionnaires to assess consumption patterns along the GCC countries.

From all the metals tested, arsenic seems to be the one with less data available and given the potential hazardous effects it seems a priority to explore seafood contamination of arsenic. Cadmium poses a threat given its distribution in these waters. Shellfish and fish contain high concentrations of cadmium and arsenic. Estimated weekly intakes of these metals from shellfish are extremely high and above the Provisional Tolerable Weekly Intake. Of all countries, fish from Oman seems to be the less contaminated.

Prevention of metal toxicity in humans as a result from exposure from the consumption of seafood depends on the environment but has to be correlated with human patterns of consumption. Nevertheless, a major public health concern for the general population could be the potential health effect of chronic low-level metal exposure that may result from modest to several servings per week of fish consumption.

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